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COMPLETE SPECIFICATION.

Improvements in or relating to Electrodes for Electrochemical Devices.

We, LEESONA CORPORATION, a corporation organized and existing under the laws of the Commonwealth of Massachusetts, United States of America, of 333, Strawberry Field Road, Warwick, State of Rhode Island, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to improved electrodes for use in an electrochemical device such as a fuel cell and to the electrochemical device employing the novel electrodes. More particularly the invention embraces a lightweight electrode comprising a porous metal support and a uniform mixture of a hydrophobic polymer and a particulate metallic substance, this metallic substance being cobalt, a mixture or alloy of cobalt and nickel, or cobalt-activated nickel. The electrodes are to be used primarily in a fuel cell as the oxygen electrode. However, as will be apparent, the electrodes can be used in other electrochemical devices where similar considerations apply.

In the art, the advantages of lightweight electrodes for use in fuel cells has been recognized. These electrodes which comprise a porous metal support coated with a catalytically active material, such as a dispersion of noble metal black, and hydrophobic polymer, being extremely thin, have low internal electrical resistance and, furthermore, take up only a very small amount of space, permitting the construction of highly compact cells having a high energy to volume and high energy to weight ratio. However, up until the present time it has been found that in order to obtain the neces-

sary catalytic activity at low temperatures, the highly expensive noble metals such as platinum, palladium, osmium, and ruthenium, are necessary. These electrodes, however, are not overly reactive when employed in an electrode for use as the cathode.

Accordingly, it is an object of the present invention to provide an improved electrode primarily for use as the oxygen electrode which possesses high electrochemical activity and does not require the expensive metals such as platinum.

According to the invention there is provided an electrode for use in an electrochemical device comprising a porous metal support uniformly coated on at least one surface with a catalytic material, said catalytic material comprising a uniform mixture of a hydrophobic polymer and a particulate metallic substance, said metallic substance being cobalt, a mixture of cobalt and nickel, cobalt-nickel alloy, or cobalt-activated nickel.

The invention also provides an electrode according to the preceding paragraph disposed in an electrochemical device for the generation of electrical current comprising means for providing an oxidant to the said electrode, at least one other electrode, which is anodic during the generation of electrical current by the said electrochemical device, and electrolyte in contact with at least one surface of each of said oxidant and anodic electrodes.

The electrode according to the invention can be constructed by applying a uniform mixture of a hydrophobic polymer and a particulate metallic substance, this substance being cobalt, a mixture or alloy of cobalt and nickel, or cobalt-activated nickel

to one or both major surfaces of a porous metal support. After applying the aforesaid mixture to the support, the total structure is heated at a temperature elevated sufficiently to sinter the hydrophobic polymer. In operation in a fuel cell, the hydrophobic polymer, being uniformly dispersed throughout the structure, will control the electrolyte interface within the electrode, preventing flooding and consequent damage or loss of power.

In the construction of the electrodes, the metal support can be a metal screen, that is, gauze or mesh, expanded metal or metal felt. It is essential that the structure be electronically conductive and able to withstand the corrosive environment of a fuel cell. Suitable porous metal supports, which are preferably from 0.5 to 1 millimeters thick with the mesh size being from 50 to 150 on the American scale, are composed of nickel, copper, iron, tantalum, zirconium, gold, silver, and alloys thereof. Primarily from the standpoint of their exceptional resistance to the corrosive environments of the cell and their relative inexpensiveness, nickel, titanium, zirconium and tantalum supports are preferred.

The polymer which is dispersed with the cobalt, cobalt and nickel, cobalt-nickel alloy or cobalt-activated nickel particles which is applied to the porous support must be relatively hydrophobic. Exemplary polymers include polystyrene, polyethylene, polytrifluoroethylene, polytetrafluoroethylene, polyvinylfluoride, polyvinylidene fluoride, polytrifluorochloroethylene, and copolymers thereof. However, because of its exceptionally hydrophobicity, as well as its resistance to heat and the corrosive environment of the electrolyte, polytetrafluoroethylene is preferred.

The mixture of cobalt, or cobalt and nickel, or Co-Ni alloy, or cobalt-activated nickel particles and polymer can be applied to the metal support screen by numerous methods with it only being essential that the polymer be uniformly distributed throughout the catalytic layer. Thus, the mixture can be applied to the metal support as a dispersion of the metal particles and the hydrophobic polymer in a suitable medium such as water. After the catalytic metal and polymer particles are mixed in the water to obtain a uniform suspension, the water is removed sufficiently to give a doughlike mass which is applied to the metal support by brushing, pressing, or rolling. Alternatively, the catalytic metal-polymer layer can be applied to the support by spraying from an aqueous dispersion. Preferably, the coated support is dried by contacting the structure with a flow of warm air and thereafter lightly pressed or rolled. The structure is then heated at an

elevated temperature to bond the catalytic layer of the metal support.

The ratio of polymer to catalytic metal in the dispersion is not critical. Normally, the desideratum is to have as light a load of the nickel and cobalt as possible but with a high surface area exposed for electrochemical reaction. In the usual construction, the catalytic metal polymer mixture will contain from about 90 to 55 percent metal and from 10 to 45 percent polymer on a weight basis. The optimum percentage is from about 80-90 percent metal and from 20-10 percent polymer on a weight basis.

Although the heating of the electrode structure at elevated temperature to bond the polymer particles together and to the metal support is essential to obtain an electrode with high mechanical stability, the temperature of the sintering and the time of the operation can vary over a substantial range. Thus, normally, the temperature of the sintering operation will be from about 180 to 325°C. for a period varying from about 5 to 45 minutes. Inasmuch as there is a direct relationship between time and temperature, within limits, if the temperature is increased, the time of the sintering operation can be reduced. It has been found, however, the greater reproducibility is obtained if the temperature is maintained between 220 and 300°C. for periods of about 5 to 35 minutes. Furthermore, as noted hereinbefore, it has been found that prior to the sintering operation it is desirable, although not absolutely necessary, to dry the electrode in air at slightly elevated temperatures, i.e., 50 to 95°C. and lightly press the structure. The sintering operation and the drying operation can be carried out in a conventional draft furnace.

Having described the invention in general terms, the following example is set forth to more particularly illustrate the invention. Parts are by weight unless otherwise specified.

EXAMPLE

A dispersion of cobalt and nickel particles at a one to one ratio on a weight basis, and polytetrafluoroethylene (PTFE) was prepared by suspending the materials in water employing a minor amount of surfactant. On a weight basis the suspension comprised 15 percent PTFE and 85 percent cobalt and nickel particles. After obtaining a uniform suspension, the mixture was sprayed on both major surfaces of a 100 mesh nickel screen, that is, gauze, having a wire diameter of 0.002" and a weight of 14.4 milligrams/cm². The structure was lightly dried in a draft furnace at a temperature of 85°C. for a period of ten minutes. Thereafter the structure was rolled lightly. After rolling, the electrode was

placed in a draft furnace and heated to a temperature of 250°C. and held for 6 minutes. The sintering served to bond the polymer particles to each other and to the support screen.

The electrode so formed was tested in a half-cell as a cathode and fed with air at 150°C. The electrolyte was a 65 percent aqueous solution of potassium hydroxide. The electrode provided data as follows:

Electrode Voltage Volts	Current Density of Electrode ma/cm ²
0.98	100
0.94	200
0.91	300

In the above example, numerous modifications can be made in the novel electrodes of the present invention. The metal support screen can be replaced with other metal supports including copper, silver, gold, iron, and platinum. Additionally, the hydrophobic polymer can be replaced with other polymers, including polystyrene, polyethylene, polytrifluoroethylene, polyvinylfluoride, polytrifluorochloroethylene, and co-polymers thereof. The cobalt-nickel catalyst can be replaced with cobalt, cobalt activated nickel particles, or a cobalt-nickel alloy. Moreover, while it is indicated that the electrode is to be used as the cathode, the electrodes of the invention can be employed as an anode and provide useful current densities. The electrodes of the invention can be employed with any of the prior art electrolytes including the alkali metal hydroxides, alkanolamines, sulfuric acid, phosphoric acid, and virtually any material which remains invariant under the operating conditions of the cell and which will provide sufficient ionic conductivity.

WHAT WE CLAIM IS:—

1. An electrode for use in an electrochemical device comprising a porous metal support uniformly coated on at least one

surface with a catalytic material, said catalytic material comprising a uniform mixture of hydrophobic polymer and a particulate metallic substance, said metallic substance being cobalt, a mixture of cobalt and nickel, cobalt-nickel alloy, or cobalt-activated nickel.

2. An electrode as claimed in Claim 1, wherein the hydrophobic polymer is polytetrafluoroethylene.

3. An electrode as claimed in Claim 1 or Claim 2, in which the metallic substance is a mixture of cobalt and nickel.

4. An electrode as claimed in any of Claims 1—3, in which the porous metal support is composed of nickel, copper, iron, tantalum, zirconium, gold or silver, or alloys thereof.

5. An electrode as claimed in claim 1, substantially as hereinbefore described.

6. An electrode as claimed in any of Claims 1 to 5 disposed in an electrochemical device for the generation of electrical current comprising means for providing an oxidant to the said electrode, at least one other electrode, which is anodic during the generation of electrical current by the said electrochemical device, and an electrolyte in contact with at least one surface of each of said oxidant and anodic electrodes.

7. An electrochemical device as claimed in Claim 6, in which the electrochemical device is a fuel cell.

8. An electrochemical device as claimed in Claim 6 or Claim 7, in which the oxidant is oxygen.

9. An electrochemical device as claimed in any of Claims 6 to 8, in which the oxidant is air.

10. An electrochemical device as claimed in Claim 6, substantially as hereinbefore described.

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